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TECHNICAL AND ENGINEERING FEASIBILITY STUDY OF THE VITRIFICATION OF PLUTONIUM-BEARING SLUDGES AT THE KRASNOYARSK MINING AND CHEMICAL COMBINE BY MEANS OF MICROWAVE HEATING

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ABSTRACT

This engineering feasibility study compared three possible technical options and their economic viability of processing plutonium-bearing sludges containing 0.6 MT of weapons-grade Pu accumulated at the Mining and Chemical Combine (MCC) at Krasnoyarsk.

In Option 1, the baseline, the sludges are processed by extraction and purification of plutonium for storage using existing technologies, and the non-soluble radioactive residues generated in these processes undergo subsequent solidification by cementation. Options 2 and 3 involve the direct immobilization of plutonium-bearing sludges into a solid matrix (without any Pu extraction) using a microwave solidification process in a metal crucible to produce a glass, which is boron-silicate in Option 2 and phosphate glass in Option 3. In all three options, the solid radioactive waste end products will be placed in storage for eventual geologic disposal.

Immobilization of residual plutonium into glass-like matrices provides both safer storage over the lifetime of the radionuclides and greater security against unauthorized access to stored materials than does the extraction and cocentration of PuO_2, supporting our efforts toward non-proliferation of fissile materials. Although immobilization in boron-silicate glass appears now to be marginally preferable compared to the phosphate glass option, a number of technical issues remain to be assessed by further study to determine the preferable immobilization option.

I. INTRODUCTION

The selection of an approach to spent nuclear fuel depends upon many factors: economic, political, social, and technical. Some countries (e.g., Canada, Sweden, US) consider it expedient to leave the plutonium in the spent fuel and place it in long-term storage for eventual disposal in deep geologic formations. Other countries (e.g., Belgium, France, Germany, Japan, Russia, Switzerland, Great Britain) consider the plutonium to be a valuable resource, and reprocess spent fuel to recover Pu for reuse.

Liquid and solid radioactive wastes containing long-lived radionuclides result from this processing, particularly for the production of mixed U and Pu oxide fuel for power stations. Considering their specific contribution to the total number of α-radionuclides, the most hazardous are transuranium nuclides such as ^{239}Pu, ^{240}Pu, ^{241}Am, and $^{242,244}Cm$. Because of the high radiation levels and long half lives, these radionuclide products must be conditioned, containerized, and disposed of using multi-barrier protection systems in order to remove them from the field of human activity for the required length of time. However, the procedures and processes required, especially for the safe geologic disposal of ^{239}Pu, have not yet been adequately developed in Russia.

Disposal and storage options (e.g., deep wells, mine-like storage in bedrock), as well as the content and form of the radionuclides, must also be considered in the evaluation of an immobilization technology. Leaching rates, radionuclide migration, control of criticality, heat release, and other factors that affect the probability of radionuclide escape from repositories have or are being studied in a number of new generation, underground laboratories placed in various geological formations: clay (Belgium), tuff (Nevada), salt mines (Germany), and granite (Canada, Switzerland).

It is very desirable to reduce the volume of these radioactive wastes and convert them to chemically and radioactively stable forms. Different types of melters and heating techniques have been explored by various countries. A number of vitrification materials have been studied. Glasses and ceramics are the primary immobilization choices for both α–nuclide and high-level wastes (e.g., nepheline-syenite aluminum silicate, lead-iron-phosphorus, titanate, monacite, natrium-zircoium-phosphate to name a few) but only the boron-silicate glass produced by induction heating in a metal crucible and ceramic melters and the aluminum-phosphate glass produced by direct heating have been used to vitrify high level waste (HLW).

In developing various processes for immobilizing HLW, it can be advantageous to fractionate the waste into a relatively short-lived Cs/Sr fraction and long-lived actinides. The short-lived fraction requires some thousands of years of retention but the second fraction is potentially hazardous for millions of years. Although glass is suited for retention of the Cs/Sr fraction, the long-lived fraction can be immobilized with those crystalline materials that are thermodynamically stable, and which consist of minerals whose stability has been reliably demonstrated over millions of years by geological data.

II. DESCRIPTION OF WORK

The principal technological flow diagrams and processes and storage equipment layouts for the comparison of immobilization technologies with the baseline PuO_2 extraction operation (Option 1) were developed using existing facilities at K-26 to the maximum extent possible to minimize costs and implementation schedules. We designed approaches to packaging, handling, storage, and geologic disposal of solidified Pu-containing sludges, and developed baseline approaches for technological handling of the solidified sludge. In addition, we listed primary technological and handling equipment; developed baseline approaches to auxiliary and service systems; and formulated principles for radiation, nuclear, and general safety.

Preliminary quantitative analyses of personnel radiation exposures during routine operations and in postulated emergencies were made for three options. We considered preliminary approaches to geologic disposal of the solid wastes, including thermal and physical calculations. To substantiate the solidified waste storage mode, we calculated the dependencies of the waste heat releases. Technical and economical data were obtained relative to options of handling the sludge, and costs broken down into separate life-cycle phases of investment and operating costs, including processing, storage, and geologic disposal.

A. Options for Immobilization

The three options considered for the immobilization of plutonium-bearing sludge at MCC are outlined in Fig. 1.

- Option 1. Sludges are processed into reusable PuO_2 with subsequent cementation of the non-soluble radioactive residues according to the existing technology for Pu extraction and purification.
- Option 2. Direct immobilization of Pu-bearing sludge (w/o plutonium extraction) into a solid glass matrix by microwave solidification in a crucible with the production of boron-silicate glass (KRI technology).
- Option 3. Direct immobilization (w/o plutonium extraction) into a solid glass matrix by microwave solidification in a crucible with the production of phosphate glass (VNIINM technology).

B. Retrieval and Preparation of Pu-bearing Sludge

The concentration of the solid phase of the sludge is 2-5 wt% for all three options. Retrieval was done with newly developed equipment, and involved the following activities:

- Agitation and retrieval of sludge with hydromonitors and hydroelevators and pulsating pumps.
- Accumulation and condensing sludge in tanks, decanting the solution into storage tanks.

In Option 1, salts in the sludge were washed out to reduce salt down to 50 g/l sodium nitrate with 1:1 ratio of sludge to condensate, evaporating the washing solution to reduce sodium nitrate from 100 to 300 g/l; underground borehole disposal of the washing solution condensates. Then the plutonium is extracted and purified. Pu is re-extracted

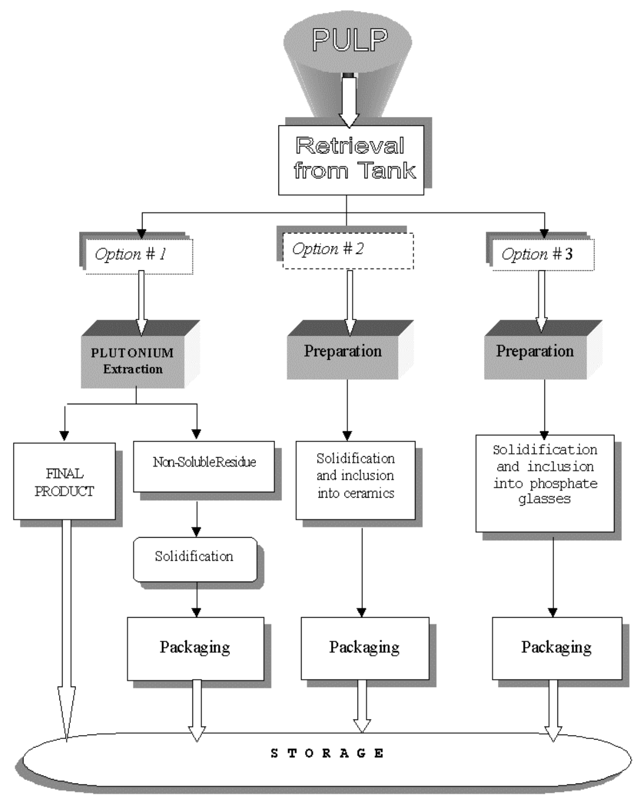

Figure 1. Engineering feasibility study options block diagram.

and fed into the PuO_2 production process, and plutonium oxide is delivered to temporary storage or shipped to a user. The non-soluble radioactive residues are cemented in sealed barrels, which are placed in protective containers and stored until geologic disposal is available.

In Option 2, the salt content was reduced to 15 g/l sodium nitrate with 1:2.6 ratio of sludge to condensate, evaporated to reduce sodium nitrate from 50 g/l to 300 g/l, underground borehole disposal of condensate, and the solid phase concentrated from 60 to 120 g/l. A dynamic filter was also used to concentrate the solid Pu phase from 60 to 120 g/l for Option 3.

C. Sludge Solidification by Microwave Heating

1. Options 2 and 3 [Vitrification]. Figure 2 is the schematic flow and material balance diagram of Pu-bearing sludge vitrification for this two-stage solidification process. The following operations are performed:

- Fluxing condensed sludge with glass-producing compositions in tanks
- Dehydrating prepared fluxed sludges at first stage of vitrification
- Separating dehydrated sludge from gas-vapor mixture and feeding the mixture to the purification area
- Solidifying dehydrated sludges in capsule-crucible at 2nd stage of vitrification
- Filling, sealing, and transporting the containers to the long-term storage.

Figure 3 shows the layout of the vitrification facility.

The gas-vapor mixture that results from sludge dehydration is cooled and condensed, undergoes a two-stage filtration and sorption process, followed by the collection of liquids with any over-the- limits of radioactive materials for deep borehole injection or other approved disposal and venting of the purified gases through a high rise stack.

One main difference between the two immobilization options that can impact the engineering results is operating temperature. Phosphate glass assumes temperatures of up to 950°C and boron-silicate glass 1250°C. In general, the higher the processing temperature, the greater are the engineering challenges. Other differences are summarized in the conclusions section.

D. Engineering Approaches to Storage of Solidified Sludge and Thermal Analysis

The MCC Radiochemical Plant has a large complex of underground cavities that are used for many purposes, including temporary storage. To provide long-term storage of waste packages (containers with vitrified waste and barrels with cemented waste) for 30 years or more, it would

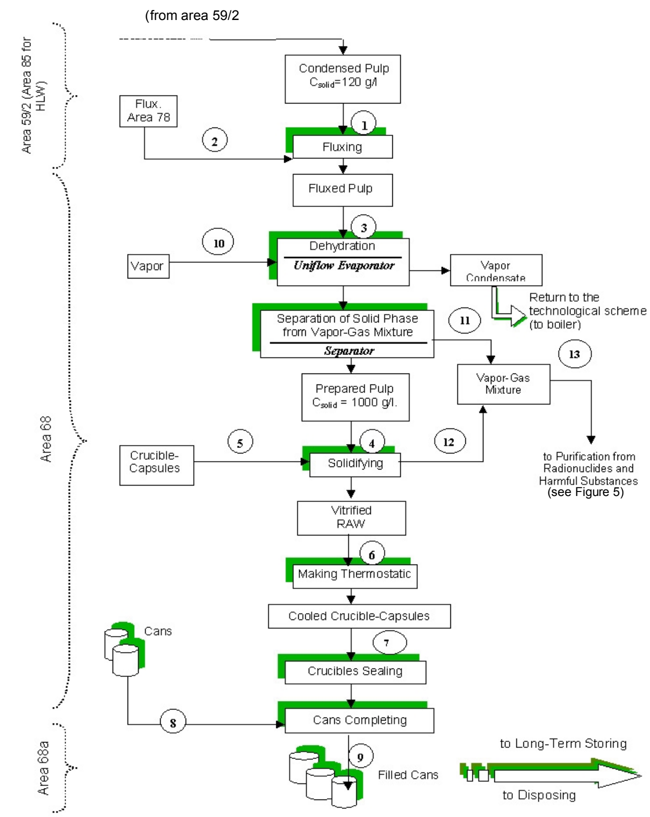

Figure 2. Vitrification process for Pu-bearing sludge (Options #2 and #3).

be necessary to modify the configuration of an existing tank facility by subdividing the tank into cells for individual packages, leaving the central channel free for observations and pumping out water in emergencies. Waste containers are loaded by a remotely controlled crane. Heat is removed via forced air, which is filtered before release to the stacks. The pressure differential inside the storage tank prevents radionuclide release to the environment via the ventilating system. The capacity and configuration of the storage facility depends upon the option selected for solidification and the corresponding size and number of packages of radioactive waste produced from processing the Pu-bearing sludge over an estimated period of 13.2 years. Figure 4 shows the layout for Options 2 and 3. The layouts are similar for all three options except for size differences in containers.

In Option 1, the total number of radioactive waste (RAW) packages produced is estimated at 9883 barrels requiring 430 pillars and 5 subdivided tanks; for Option 2, it is 6800 cans, 284 pillars and 2 tanks; and for Option 3, it is 11,860 cans requiring 492 pillars and 3 tanks.

The specific heat release of radioactive waste solidified according to the three options is given in Table 1.

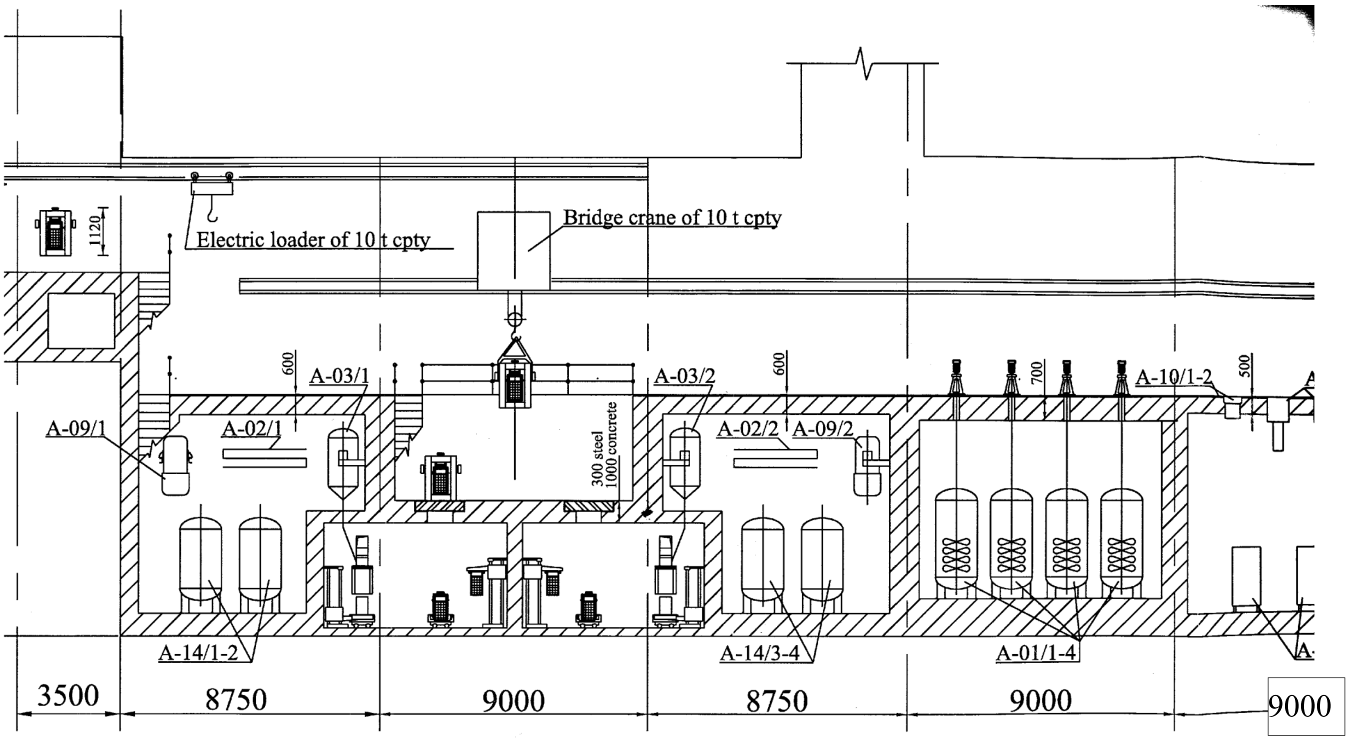

Figure 3. Layout of vitrification facility.

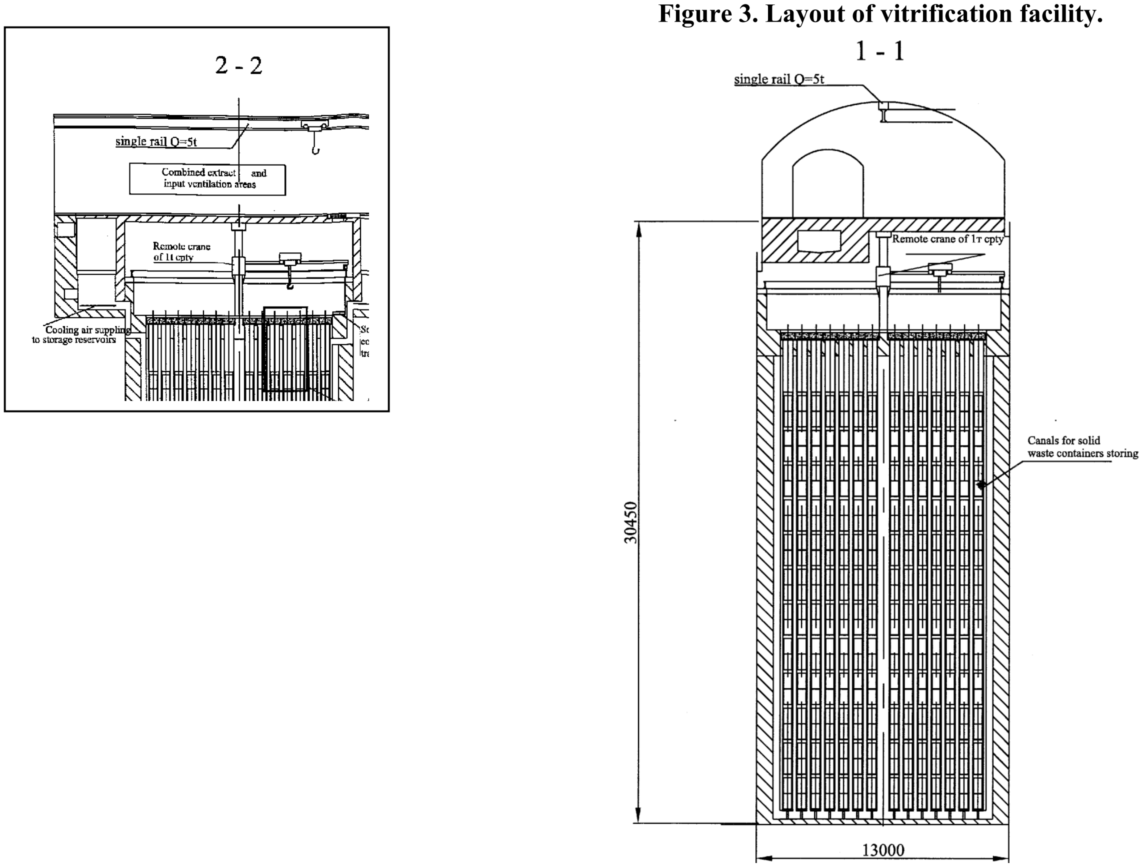

Figure 4. Layout decisions, sections 1-1, 2-2 for area 86a, Options 2 and 3.

Table 1.Variation of RAW heat release in time, W/dm^3.

Time (y)	Conditioned waste from processing (W/dm^3)		
	Option 1 (baseline Pu recovery)	Option 2 (boron-silicate glass)	Option 3 (phosphate glass)
0	$7{,}0\cdot10^{-3}$	1,0	0,59
10	$1{,}1\cdot10^{-3}$	$1{,}5\cdot10^{-1}$	$8{,}8\cdot10^{-2}$
20	$9{,}1\cdot10^{-4}$	$1{,}2\cdot10^{-1}$	$6{,}9\cdot10^{-2}$
30	$7{,}4\cdot10^{-4}$	$9{,}6\cdot10^{-2}$	$5{,}5\cdot10^{-2}$
Factor of increase in RAW heat release for 30-y storage = -10.			

The thermal mode of the storage facility was estimated with a computer model that calculates a conjugated heat exchange between tubes packed with radioactive waste containers and cooling air under forced ventilation. The model allows for variations in the waste heat release in the course of radioactive decay. A number of conservative assumptions were made (e.g., calculating that containers were uniformly filled 100% with radioactive waste when actually the crucibles are 80% filled with vitrified waste and barrels 70% with cemented waste, only cooling air from below was considered, and the facility treated as completely filled). The results show that, for safe operation, the temperature must be less than 45°C regardless of the option selected for processing the sludge. To achieve this temperature, cooling air at 25°C must be supplied at a rate of 0.1 m/s for the boron-silicate glass and at 0.02 m/s for packages of phosphate glass or cemented waste.

E. Analysis of Radiation Impacts from Processing Plutonium-bearing Sludges

The immobilization of plutonium-bearing sludges is associated not only with radioactive impact on personnel and the environment but also with the possibility of reaching, under particular hypothetical conditions, nuclear criticality characterized by both radiation (fission products) and mechanical (explosion) impacts. Therefore, safety provision principles shall meet requirements for both radiation and nuclear criticality safety.

Monitoring systems are in place at the sludge processing and solid waste storage areas located on the site of an operating radiochemical plant and at monitoring zones outside the plant established by radiation safety regulations. Releases of radionuclides and harmful chemical substances with gas and aerosols are monitored continuously. At territory adjacent to the MCC, the content of radionuclides in the atmosphere, precipitation, soil, and plants is monitored at a distance of 15 km from the release source. The Yenisey River water is subject to control to 15 km and fish controlled at a distance of up to 50 km downstream from the discharge channel. Within the MCC's health protection zone, it is possible to detect ^{137}Cs, ^{144}Ce, ^{106}Ru, and ^{60}Co; the average annual volumetric activity of these radionuclides is under allowable limits. In the atmosphere next to the MCC, mostly ^{137}Cs is found, with activities less than the allowable regulated values. At all monitoring points, the gamma-radiation absorbed dose rate is equal to 7-12 μR/hr. The dosimetric survey of 23 places located at distances up to 1500 km downstream from the combine discharge channel shows that the exposure dose rate there does not exceed 14 μR/hr. Agricultural products in the regions of the floodplain of the Yenisey River most subject to radionuclide accumulation have contents of ^{137}Cs, ^{90}Sr, and ^{65}Zn that are 150-600 times lower than permissible. At present, average radiation doses at operational radiochemical facilities are equal to 0.04-0.75 cSv with a norm of 2 cSv. Nuclides in some ventilation releases are 0.230-0.652% of the maximum allowable value. The gamma background within the monitoring zone is the same as natural and equal to 10-18 μR/hr. The annual exposure dose of population and critical group of population is 4-40 μSv, less than the requirements.

The nuclear safety requirements must prevent criticality during normal operation and in case of design-based accidents. The technology for immobilizing Pu-bearing sludges is based on the concept of providing nuclear safety by utilizing nuclear safe equipment in which the safe concentrations of plutonium or safe allowable fissile mass is not exceeded (as well as maintaining the technological parameters that provide the safe volumetric concentrations of plutonium in technological equipment). Safe and allowable parameters of particular equipment will be determined at the subsequent design phase based on the value K_{ef} not exceeding 0.95. The margin factors are set forth in Russian requirements.

General safety requirements are also specified for fire protection, lighting, ventilation, electrical safety, personnel protective equipment, and working conditions.

Contamination and dose control during sludge and Pu retrieval and sludge vitrification are strictly controlled. The production facilities are divided into three areas:

- Area 1. Technological equipment and communications area. Personnel access prohibited.
- Area 2. Servicing area for equipment repair, etc. All personnel activities in Area 2 are performed with authorized access and under observation of the radiation security control system. Personnel must use respirators.
- Area 3. Permanent personnel stations. Sanitary sluices mounted between areas 2 and 3 must meet regulatory requirements.

The radiation level of technological processes is controlled by signal measuring devices with range of 0-1000 μR/s and detection threshold of 0.3 μR/s. There are 23 check points in Area 2 that cover all of the processing points. A stationary air test system is used to control activity of radionuclides in work areas. The following items are controlled according to a schedule of planned measurements:

- Exposure dose capacity in work areas 2 and 3;
- Facilities and equipment contamination with α-, β-, and γ-nuclides;
- Activity capacity of the working area atmosphere contaminated with α and β nuclides;
- Individual radiation monitoring of personnel outer contamination with individual radiation monitors, cassettes processed monthly;
- Individual radiation monitoring during service and repairs;
- Clothes, shoes, and skin contamination of personnel during approved schedule and during activities requiring authorized access.

All personnel make a complete change of clothes in the sanitary booth when entering. Upon exit, contamination is measured in the booth after shower body contamination is measured and individual clothes contamination monitored at the exit from the booth. Annual personnel dose does not exceed 20 mSv. Radiation monitoring is done by a worker with a specific qualification. Service and repair of equipment and radiation monitoring devices requires a worker qualified as a specific mechanic of control and monitoring equipment and automatic machinery.

Plutonium retrieval, in addition to the requirements above, requires a radiation monitoring technician with 900 hours experience and a specific qualification. Service and repair of radiation monitoring control equipment requires 950 hours of work experience by a mechanic of control and monitoring equipment and automatic machinery with a specific qualification.

Sludge vitrification requires 1000 hours of work experience and a specific qualification for radiation monitoring control and 900 hours of work experience and a specific qualification of mechanic of control and monitoring equipment and automatic machinery

A list of emergencies was generated and assessed for the options but is not presented here.

F. Site Selection and Geologic Disposal of Solidified Plutonium-containing Waste

Ancient metamorphous beds of Kansk series, granitoids of Tatarsk complex, granites of Belogorsk massif and some other geological formations were considered as a possible environment for isolation of solidified radioactive waste. The comparative analysis of the sites was based on the following:

- Determination of the geological formation based on geological routes,
- Study of natural and artificial outcrops;
- Analysis of tectonic breakup and particularities of newer and modern tectonics;
- Analysis of fractures by techniques of on-ground electro-exploration;
- Analysis of inner structure of a site with available geophysical techniques.

As a result, the Nizhnekansky massif of granitoids was singled out as the more promising region for deep borehole disposal of radioactive waste. This massif is one of the biggest granitoid massifs of Middle Siberia. Its outcropped area is more than 1500 km^2 (3500 km^2 given the buried portion of the massif). This very thick massif consists chiefly of biotite granites and granodiorites; its probable age is 850 ± 60 million years. The northern-western border is located 3 km from the MCC plant site.

To select promising sites within the massif, various techniques for estimating the tectonic faults, newer and modern tectonic activities of the region, and geochemical appropriateness of granitoids of Nizhnekansky massif were applied. Thirteen potential sites were identified and narrowed to two sites, later named «Itatsky» and «Kameniy». Electro-exploration confirmed that these sites have monolithic granitoid blocks. Two options were reviewed in this feasibility study: (1) disposal in a mine in horizontal rock cavities at a depth of more than 500 m, and (2) disposal in deep boreholes of diameter sufficient to emplace one shaft to a depth of 1000-2000 m.

1. Mine Geologic Disposal. The mine option provides for three shafts, a number of horizontal cavities located not deeper than 500 m, wells with a diameter suitable for one tube and equal to some tens of meters in depth, and transportation and auxiliary cavities intended for repository uses. Shaft 1 is the technological shaft for descending with radioactive waste. Shaft 2 is for bringing people, material, and equipment up or down, and for removing excavated rock when constructing cavities. Shaft 3 is for ventilation, and serves as an emergency exit to the surface from the underground cavities. The on-ground complex in the mine option consists of:

- Technological building where packages are subject to reception, transportation, technological and other activities, and transferred to the underground complex located under Shaft 1;
- Buildings above Shafts 2 and 3;
- Engineering and auxiliary buildings.

2. Deep Borehole Disposal. The deep borehole option provides for establishing a network of boreholes. The boreholes are drilled with a separation between them for

radioactive waste disposal that is based on mining and geological conditions and allowable thermal effects of the radioactive waste on the rock massif. The borehole design comprises (1) a head through which containers with radioactive waste are transferred, (2) a grouted portion of the borehole (in fracturing dehydrated zone of the rock massif), and (3) a working portion thereof.

The on-ground complex in the deep borehole option comprises:

- Technological buildings located above the head of each borehole where packages are subject to reception, transportation, technological and other activities, and transferred to the borehole head;
- Engineering and auxiliary buildings.

3. Analysis. The analysis shows that it is not economically expedient to establish a deep borehole disposal in geological formations for one small group of HLW-containing long-lived elements more related to the mine option of disposing. Therefore, it is not expedient to create a complex of buildings and mine cavities to solely dispose of only solidified plutonium-containing pulps. We recommend a multi-use mixed source repository be established to dispose of other types of waste (e.g., solidified HLW and nuclear spent fuel which is not subject to reprocessing) in order to share infrastructure and other costs.

The deep borehole option does not require that a big underground complex be established (i.e., the number of boreholes to be drilled is the number needed to dispose of solidified plutonium-containing pulps; see Table 2). It is more reasonable to consider this facility if Pu-containing pulp wastes are the only type of wastes being disposed of in a geologic site.

Along with disposal in deep bedrock, the Engineering Feasibility Study also analyzed the possibility of immediate disposal of the cemented non-soluble residues from the Pu-bearing sludge process in storage areas located in rock cavities at MCC. Pursuant to the classification of solid waste that is valid in the Russian Federation, after 30 years' storage, the cemented waste may be regarded as Intermediate Level Waste (A_{spec} = 0.8 10-1 Ci/dm^3) and a concentration of plutonium in the waste is equal to 20 mg/kg.

The proposed procedure is to store the solidified waste for 30 years subject to final disposal in the same tanks. For this purpose, the structure is to be made monolithic by filling the inter-tube space with the specially designed cement solution that is resistant to radiation, temperature, and water, and the tube channels are to be bound by welding. In this case, a firm and stiff structure should be built. To eliminate consequences of a possible off-gassing, the gaseous phase output may be provided for. The final conclusion on such a technique of disposing of the waste is to be considered separately, and safety analysis is to be carried out in detail.

Table 2. Boreholes required to dispose of conditioned RAW under three processing options.

Option	Amount of RAW to be disposed of		
	m^3	No. of packages	Boreholes required
Option 1. Existing process for sludge treatment with Pu extraction and purification, and cementation of non-soluble residues	1571	9834 barrels	17
Option 2. Microwave solidification of Pu-bearing sludge producing boron-silicate glass	544	6800 cans	15
Option 3. Microwave solidification of Pu-bearing sludge producing phosphate glass	949	11860 cans	26

To provide rough relative estimates of disposal expenses, the Engineering Feasibility Study considered only deep borehole disposal using the following parameters for all three options of waste handling:

Burial site operating lifetime	5 years
Borehole depth	1000 m
Burial depths	1000 – 500 m
Borehole diameter	920 mm
Type of a composition to isolate RAW in wells	Bentonite
One borehole capacity	454 barrels or 581 cans
Borehole separation	50 m
Area to locate the boreholes	60 000 to 80 000 m^2

4. Transportation and Technological Geologic Disposal Plan. Upon expiration of the storage time, RAW will be transported by rail in protective containers and packages (TK-6 or others) from the storage facility over a 40-km railroad to an above-surface building at the geologic disposal site. One railway vehicle will carry containers. Two transportation containers will provide continuous operation: one for loading into the storage facility, the other for unloading at the above-ground complex.

The deep borehole disposal transportation and technological scheme involves the following operations:

- Taking RAW barrels/cans out of the MCC storage facility and placing them into a protective container with available devices;
- Transportation of the protective container to the completion chamber;
- Completion of the transportation container;
- Placing on the special railway vehicle;

- Transportation of the containers to a technological building at the borehole head and entering into a railroad passage of the building;
- Disintegration of the transportation container;
- Placement of cans/barrels on a prepared borehole mouth, i.e. filled with bentonite solution up to a required level. The borehole mouth is equipped with a gate preventing random releases of radioactive gases from the borehole to the room;
- Descending the cans/barrels into the borehole with the specially developed equipment like that used for oil well maintenance (e.g., KORO, 80 MT lift capacity). A special vessel with bentonite paste to fill a space between the barrels is lowered in the same way;
- Closing the borehole mouth and returning the empty container to the reloading chamber to load the next can/barrel.

5. Storage Facility and Geologic Disposal Site Safety Analysis. Three mathematical models were used to estimate the safety of the storage facility and burial site. A one-dimensional model of radionuclide diffusion in a non-uniform environment calculated the concentration at any time at any distance from the source and considered the sorption, dispersion, and radioactive decay of radionuclides. The second model was of the distribution of radionuclides in a geologic fracture to determine radionuclide concentrations at any distance from the source and estimate the level of fracture contamination. The third model estimated dose rates by calculating total dose of inner radiation in five food chains based on radionuclide concentrations in water. Initial concentrations, activities, and migration characteristics of radionuclides were considered.

Five scenarios were postulated; four for the geologic disposal site and one for the storage facility. Calculations were carried out in two phases. First, the maximum concentration of radionuclides at the outer border of the disposal site or storage facility were determined. Second the radionuclide migration in rock fractures and the maximum distance from the site that the radionuclides could cover was determined. After completing all calculations for the five scenarios, the worst case was selected and the radiation dose calculated.

The worst case radioactive release most likely takes place when the steel shroud surrounding a barrel with cemented waste is completely corroded and radionuclides migrate from cemented waste through the bentonite to the surface through the rock fracture (lineament). For this case, dependencies of a total internal dose on time in each radionuclide have been calculated based on five food chains. The check point is a point of possible outcrop of the lineament at the daylight surface. The radionuclide concentration at this point was the basis for calculating the radiation doses.

The ^{137}Cs dose is continuously decreasing in time. Variations of ^{90}Sr and ^{239}Pu doses have maximums (at 550 and 560000 years, respectively), and the ^{238}U dose within the calculation period (2 million years) is continuously increasing. Despite the different behavior of the dose variations, all maximum values of the doses calculated for the possible outcrop of the lineament at the day surface are *much less* than required by all valid regulations. It means that the deep borehole burial site remains safe for the environment even in the case of occurrence of the worst accidents.

G. Economic Evaluation of Pu-bearing Sludge Immobilization Options

The economic data were calculated for the three options: baseline PuO_2 production, immobilization in boron-silicate glass, and immobilization in phosphate glass. A number of conditions were present or assumed for these calculations. Among them, secondary liquid radioactive waste arising from operations for immobilization is subject to injection at the deep well disposal according to existing technology. Investments and operating expenses associated with the options for the immobilization of plutonium-bearing sludges were determined. These options were compared in terms of total costs associated with sludge handling for (1) the facility operational lifetime (13,3 years), (2) the period of long-term storage (30 years), and (3) the period of disposal at geological wells (5 years).

Economic data was calculated based on Russian legislation and relative to 1991 prices. Costs in US Dollars are based on the currency exchange rate in 1991 since that was the last year of a stable ratio of the Russian Ruble to the US Dollar. The 1991 currencies rate is equal to 1.

The economic data were calculated based on design developments assumed for main phases of technological processes for handling sludges and on Russian analogues associated with this project.

1. Economic Data on the Sludge Processing Facilities. The following activities were considered in calculating the investments required for implementation of the technical approaches to the three options:

- Purchase and installation of technical facilities (common for all options) for agitating and retrieving sludges from tanks;
- Reconstruction of facilities for washing the sludge out of sodium salts (Options #1 and #2) and for condensing the sludge (Options # 2 and #3);
- Reconstruction of the solutions evaporation facility (Options # 1 and # 2);

- Purchase and installation of equipment for cementing the sludge (Option # 1);
- Purchase and installation of two vitrification facilities (Options #2 and # 3) with the performance of construction activities to provide 7200 m^3 for placing the equipment;
- Modifications to auxiliary and service subsystems (ventilation, air supply, power supply, etc.).

Costs associated with the construction activities have been included in the calculations.

To place the required equipment for handling the sludge-like waste in existing rock cavities, extra deactivation and dismantlement activities must be carried out. It is proposed to deactivate areas prior to dismantlement of the equipment according to the existing technology with operational means and instruments.

Costs for the dismantlement activities were determined based on the minimal required dismantlement of equipment and pipelines preserving the building structures.

Options # 2 and # 3 take account of the costs for dismantlement of the cementation facility whose performance does not meet valid regulatory requirements for further use.

Table 3 gives extra expenses associated with implementation of technical approaches to handling plutonium-bearing sludges.

Annual operating costs for handling with plutonium-bearing sludges have been calculated according to valid regulatory and legislative documents. These costs include:

- Purchase of chemicals and materials including transportation and procurement expenses;
- Payments for electricity;
- Wages for service personnel and payments to social funds;
- Labor security (special food and clothes);
- Building and equipment depreciation;
- Building and equipment up-keeping;
- Injection of secondary waste at the underground repository;
- Services rendered by other MCC shops;
- General expenses;
- Taxes and payments to the industry non-budgetary funds;
- Radiochemical reprocessing to extract uranium and plutonium (Option # 1).

The building and equipment depreciation are equal to 1.1% and 8%, respectively, according to the regulations. Costs for (1) building and equipment up-keeping, (2) injecting the secondary liquid radioactive waste at the underground repository, (3) services rendered by other shops of MCC, (4) general needs, (5) wages, (6) electricity, (7) radiochemical reprocessing of the nitric acid solution to extract uranium and plutonium, and (8) taxes and payments are based on data factual to MCC.

Table 3. Initial investments in technical approaches to implementing Pu-containing sludge processing facilities ($,K).

Description	Option #1	Option #2	Option #3
1. Dismantling of the equipment and pipelines	398.0	1609.9	1609.9
2.Hardware for washing out, lifting and removal of sludge.	550.1	550.1	550.1
3. Reconstruction of unit for washing sodium salts out	7.2	7.2	-
4. Reconstruction of the unit for condensing the sludge at the filters	-	207.0	207.0
5. Reconstruction of the solution vaporization unit	78.1	78.1	-
6. Purchase and assembling of the cementing facility	3463.4	-	-
7. Purchase and assembling of the vitrification facility	-	9304.7	9304.7
8. Reconstruction of the auxiliary and maintenance systems	1198.6	1657.6	1804.0
TOTAL	5695.4	13414.6	13475.7

Given the general nature of these calculations in determining costs for the sludge solidification, the other expenses are carried at 5% of the total costs calculated. Investments associated with the disposal of the conditioned radioactive waste in deep borehole l repositories at «Itatsky» or «Kameny» sites of the Nizhnekansky massif located 25 to 30 km from Mining and Chemical Combine have been determined relative to the plutonium-bearing sludge processing in term of:

- Construction of on-ground facility;
- Drilling 920-mm diameter, 1000-m depth boreholes:
  Option #1 – 17 boreholes
  Option #2 – 15 boreholes;
  Option #3 – 26 boreholes.
- Purchase of transportation vehicles (car-platform and two car-containers of TK-6 type);
- Laying of 40-km length railway.

Annual costs associated with drilling the borehole to disposal of packages with solidified HLW were taken into consideration in the annual operating costs.

Economic data associated with the existing handling of Pu-bearing waste that involves extraction of plutonium and alternative options involving the microwave solidification of sludges without the extraction of plutonium have been compared in terms of total costs for (1) sludge processing, (2) long-term sludge storage and (3) disposal of sludge at geological wells. This comparison includes:

- Investments in establishing facilities for (1) sludge processing, (2) sludge long-term storage and disposal;
- Operating costs associated with the sludge processing for 13.2 years;
- Operating costs associated with the radioactive waste long-term storage for 30 years including waste reception (13.2 years) and disposal at the disposal facility (16.8 years);
- Operating costs for the period of 5-year disposal of the radioactive waste.

The comparison of economic data is given in Table 4.

III. RESULTS AND CONCLUSIONS

A. Summary of Results

This Engineering Feasibility Study assesses the technical possibility and economic viability of two options of immobilizing plutonium-bearing sludge accumulated at Mining and Chemical Combine and compares them with the existing baseline option.

The following activities have been carried out to obtain the objectives of this study:

- Plutonium-bearing sludges have been characterized;
- Primary characteristics of the solidified sludge (annual and total output, specific activity, radionuclide content, chemical composition, heat release) have been calculated for the following options:
  - Option # 1 (baseline): processing of the sludge according to existing technology involving extraction and purification of plutonium with subsequent cementation of non-soluble residues that arise from the process;
  - Option # 2 (Khlopin Institute): direct sludge immobilization (w/o Pu extraction) into a solid matrix using microwave solidification in a metal crucible with the production of boron-silicate glass;
  - Option # 3 (Bochvar Institute): direct sludge immobilization (w/o Pu extraction) into a solid matrix using microwave solidification in a crucible with the production of phosphate glass.

Regarding Option 1, cementation of non-soluble residues arising from extracting Pu from the sludge, this feasibility study uses data provided by MCC and design approaches developed by the Krasnoyarsk branch and Mai division of VNIPIET.

Regarding Options 2 and 3, sludge solidification w/o Pu extraction, the principle and technological flow diagrams, and storage equipment layouts have been developed. Approaches to packing the solidified sludges have been designed.

We developed baseline approaches to transportation of and technological handling of the solidified sludge and listed primary technological and transportation equipment. We also developed baseline approaches to auxiliary and service systems and formulated principles for radiation, nuclear, and general safety. Preliminary quantitative analysis of the personnel radiation exposure during routine operation and in emergency has been made. Preliminary approaches to decommissioning of the buildings and waste placement procedures have been considered (allowing for thermal and physical calculations).

To substantiate the solidified waste storage mode, we calculated the dependencies of the waste heat release.

Technical and economical data relative to options of handling the sludge have been obtained, and costs broken down into separate handling phases.

The options were compared and assessed based on our criteria for the following:

1. Process performance (e.g., temperature, volume, release of radionuclides);
2. Preliminary technological equipment and approaches to reconstructing the area;
3. Characteristics of solidified waste;
4. Safety of the solidified waste storing and disposal;
5. Economic data.

Table 5 contains a summary of the option comparisons. Although immobilization in boron-silicate glass appears now to be marginally preferable compared to the phosphate glass option, a number of technical issues remain to be assessed by further study to determine the preferable immobilization option.

B. Conclusions

Results of the work performed and analysis of the comparison criteria have shown the following:

1. Process Performance Characteristics. The baseline plutonium recovery process, Option #1, has an advantage over Options #2 and #3 in terms of development of the solidification process because it is carried out at lower temperatures and with a minimum of radionuclides generated in off-gasses during sludge solidification.

Option #3 has an advantage over Options # 1 and # 2 in terms of minimizing the secondary liquid technological wastes arising from the immobilization process, which are then subject to underground deep borehole disposal as liquids.

2. Primary Technological Equipment and Area Reconstruction. The baseline option [#1] has an advantage over both of the vitrification options because design documentation already exists for non-standardized cementation equipment. All three options are at the same level of design documentation development for non-standardized handling and technological equipment, as well as for the equipment for temporary storage of solidified products prior to geologic disposal. All three options are developed to the same extent in regard to modifications to existing areas at K-26 for installation of the appropriate equipment and for transportation of the solidified product to the temporary storage facility. Option #1 does have an advantage over Options #2 and #3 in that fewer underground tanks must be modified to establish the temporary storage facility for the solidified products.

3. Solidified Sludge Characteristics. Option #2, immobilization in boron-silicate glass, has the following advantages over Options #1and #3. Lower leaching factors for alkali earth and transuranic elements inherent in a boron-silicate glass matrix;

- Higher percentages of inclusion of plutonium and other radionuclides, which preserve the stability to ionizing and thermal impacts when stored and subsequently disposed of;
- Crystalline structure;
- Lower specific volumetric outputs of the solidified products, again due to higher percentages of inclusion of plutonium and other radionuclides; and
- Higher matrix densities.

4. Safety of Storage and Geologic Disposal. Safe storage and geologic disposal of solidified sludge requires that the cooling air temperature be maintained below 45°C for the entire time of storage. Our estimates show that to meet this requirement for boron-silicate glass, the cooling air must be supplied at a temperature of 25°C at a rate of 0.1 m/s. For phosphate glass and cement compound, it is sufficient to again supply the cooling air at 25°C but at a rate of 0.02 m/s. Thermal and physical calculations show that the radioactive waste heat released in the geologic disposal site has no practical effect on the temperature of the surrounding rock massif. An increase in temperature in the center of the disposal area containing radioactive waste will not exceed 9°C. The increase in temperature of the rocks inside the disposal site at a distance of 25 m from the disposal borehole will not exceed 2°C. Such increases in temperature inside the disposal site will not affect the rock temperature outside the disposal facility. Estimates of ecological safety of the storage facility and geologic disposal site show that storage of the solidified waste for the entire lifetime of the storage facility will generate radioactive dose rates to the exposed population below allowable rates, and the design of the deep geologic disposal site allows the safe isolation of the radioactive waste for the entire period of actual danger from the radionuclides. The distance over which a concentration of ^{90}Sr transports does not exceed 160 m, and that for ^{239}Pu does not exceed 110 m for any option.

5. Economic Data. Total investment and operating costs to implement the different technical approaches to immobilizing the plutonium-bearing sludge with the production of boron-silicate and phosphate glasses and to establish a long-term storage facility for the solidified waste are equal to 84.9 and 87.4 million US$, respectively. This is ~22 to 25% less than the total investment and operating costs for implementation of Option 1, the existing technology. Considering all costs, the production process for boron-silicate glass, Option 2, costs 15% less than Option 1 and 30% less than Option 3.

Assuming the geologic disposal of conditioned waste into deep boreholes of ~1 km, preliminary comparisons of overall economic data show that sludge immobilization in boron-silicate glass may be marginally preferable to Option 3. However, both immobilization options are more economic than Option 1, the Pu extraction baseline.

C. Discussion and Recommendations for Future Work

Immobilization of the plutonium-bearing sludges into glass-like matrixes instead of cementation matrixes increases the waste storage safety by fixing plutonium reliably in a strong and stable material. It also provides increased physical protection and nuclear safety of solidified waste during long-term storage and makes possible the final disposal of solidified waste into geological formations. Preliminary estimates show that keeping solidified plutonium-bearing materials in deep geologic disposal sites is safe. This type of disposal also excludes non-authorized access to these plutonium containing wastes, and resolves some issues associated with non-proliferation of fissile materials.

We recommend that the pre-design activities be continued into the next phase of development, the "Declaration of Intent" and the "Investment Justification." In these phases, we would further develop the processes to immobilize plutonium-bearing sludges (without any Pu extraction) into a solid matrix using microwave solidification to form boron-silicate glass (Option #2) and phosphate glass (Option #3).

Table 4. Technical and economic data on options for sludge solidification.

Description	Unit	Opt.#1 (Pu removal)	Opt.#2 (boron-silicate glass)	Opt.#3 (phosphate glass)
1. Sludge volume to be processed over entire period	m^3	7260	7260	7260
-per year	y	550	550	550
2. Mass of plutonium in the sludge	kg	593.3	593.3	593.3
3. Mass of uranium in the sludge	t	531.24	531.24	531.24
4. Solidified RAW volume: -over whole period; -a year	m^3	 1573.0 119.2	 544.0 41.2	 949.0 72.0
5. Packing tare for solidified RAW: 5.1. Capsules (steel 10; weight 86 kg; ∅ 426 mm; H=800 mm); 5.2. Solidified waste container (stainless steel; weight 15 kg; ∅ 478 mm; H=1000 mm); 5.3. Barrel (V=0,2m^3, H=850 mm, ∅ 600 mm; steel 3, weight 60 kg);	pcs/y	 – – 745	 516 516 –	 900 900 –
6. Liquid waste volume to be injected in existing borehole burial site: -over whole period: -a year	 m^3	 28248 2140	 15972 1210	 10758 815
7. Additional number of operating staff, *TOTAL* : -solidification facility; -long-term storage facility	man	 *73* 48 25	 *83* 58 25	 *83* 58 25
Invested cash for handling with sludges: 8.1 sludge processing; 8.2 long-term storing solidified sludges; *TOTAL sub-items 8.1-8.2* 8.4. establishing the geologic disposal burial complex **TOTAL item 8**	$, K	 5695.4 7194.5 *12889.9* 65958.3 **78848.2**	 13414.6 3701.8 *17116.4* 61138.7 **78255.1**	 13475.7 4796.4 *18272.1* 87654.8 **105926.9**
9. Annual operating costs for handling with sludges: 9.1 Sludges processing, 9.2 Receiving sludges for long-term storing, -including packing tare for waste -long-term storing; -solidified waste burial	$, K	 6034 1623.9 9.9 162.4 41257	 3567.5 1392.6 26.9 139.3 38330.2	 3554 1493.4 46.9 149.3 52716.6
10. Total operating costs for: 10.1 Sludge processing over 13.2 yrs; 10.2. Receiving of solidified waste for long-term storing over 13.2 yrs; 10.3. Long-term storing solidified waste over 16.8 yr; **TOTAL sub-items 10.1-10.3**	$, K	 79648.8 21435.5 2728.3 **103812.5**	 47091.0 18382.3 2340.2 **67813.6**	 46912.8 19712.9 2508.2 **69133.9**
10.4. burial of solidified waste over 5 yr **TOTAL item 10**	$, K	206285 **310097.6**	191651 **259464.5**	263583 **332716.9**
11. Comparable total costs for handling with sludges: 11.1. W/o costs for solidified waste burial 11.2. Including costs for solidified waste burial	$, K	 116702.4 388945.8	 84 930 337719.6	 87406 438643.8
12. Change in total costs: 12.1. W/o costs for solidified waste burial 12.2. Including costs for solidified waste burial	%	 100% 100%	 73% 87%	 75% 112%

Table 5. Comparison of two options for vitrification of plutonium-bearing sludges with current, baseline techniques for Pu extraction and cementation of non-soluble residues.

Item	Criterion	Baseline (Option #1)	Boron-silicate glass (Option #2)	Phosphate glass (Option #3)
1.	**Characteristics of the Process**			
1.1	Sludge solidification temperature, (°C)	20	1250	950
1.2	Amount of secondary technological liquid waste subject to underground disposal, (m3/yr)	2140	1210	815
1.3	Escape of radionuclides into the gas purification system (Ci/yr)	$1.5 \cdot 10^{2}$	$7.7 \cdot 10^{4}$	$7.7 \cdot 10^{4}$
2.	**Primary Technological Equipment and Reconstruction Approaches**			
2.1	Primary technological equipment for solidification of the sludge:			
	-Availability of design documentation on non-standardized equipment of the pilot facility	Design documentation has been developed	Subject to development	Subject to development
	-Laboratory or pilot specimen of the facility is available	Pilot specimen of the facility is available	Laboratory equipment is available	Laboratory equipment is available
2.2	Primary transportation and technological equipment:			
	-Availability of design documentation on non-standardized equipment	Design documentation on a container for transporting a barrel with solidified sludge	Subject to development	Subject to development
	-Availability of pilot specimens	Not available	Not available	Not available
2.3	Primary technological and transportation equipment for the temporary storage facility			
	-Availability of design documentation on non-standardized equipment	Subject to development	Subject to development	Subject to development
	-Availability of pilot specimens	Not available	Not available	Not available
2.4	Necessity to modify area 86a to establish a temporary storage facility for solidified waste	5 tanks	2 tanks	3 tanks
3.	**Solidified Sludge Characteristics**			
3.1	Form of the solidified waste	Cement compound	Boron-silicate glass	Phosphate glass
3.2	Content of plutonium, mg/l	~ 60	~ 1100	~ 620
3.3	Specific activity, Ci/l	1.78	265	152
3.4	Amount of the solidified sludge, m3	1573.0	544.0	949.0
3.5	Specific heat release, W/l	< 0.007	1	0.59
3.6.	Leachability, g·cm-2··day-1	Cs, Sr $\leq 10^{-3}$	Cs $\leq 10^{-6}$, Sr $\leq 10^{-7}$, Pu $\leq 10^{-8}$	Cs $\leq 10^{-5}$, Sr $\leq 10^{-6}$, Pu $\leq 10^{-7}$
4.	**Safety of Solidified Sludge Storage and Geologic Disposal**			
4.1	Excess of the surrounding massif's temperature over geothermal one, °C	Insufficient	< 9	< 9
4.2	Rate of air feed to the storage facility, m/s	> 0.02	> 0.1	> 0.02
4.3	Maximal advance of the concentration from a solidified waste container along with fracture in rock, m	< 160 (for ^{90}Sr) < 110 (for ^{239}Pu)	0.2	0.2

The following technical issues also need to be assessed in these phases:

- Optimization of the size of the crucible;
- Optimization of the procedure to place solidified sludge in the storage facility;
- Specification of technological performance of the sludge solidification in terms of nuclear safety, including that for real sludge and simulations using pilot facilities placed in hot cells at MCC;
- Development of technology and equipment operating modes, including testing of serviceability of remotely controlled devices to replace the equipment and handle packages with solidified sludge on a pilot facility;
- Development of design documentation for non-standardized technological and transportation equipment;
- Study of solidified sludge storage and disposal safety by applying mathematical models to obtain risk estimates;
- Determination of an optimal amount of plutonium in solidified waste subject to disposal;
- Study of physical and chemical properties of the glasses and their components when subjected to long-term radiation exposure;
- Impact of temperature and underground water; and
- Development of regulatory documents addressing issues associated with storage and disposal of plutonium-bearing materials.

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